UCRL-LR-130250

Radionuclides in Sediments and Seawater at Rongelap Atoll

V.E. Noshkin
W.L. Robison
R.J. Eagle
J.L. Brunk

March 1998

Lawrence Livermore National Laboratory

Work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract W-7405-ENG-48.

UCRL-LR-130250
Distribution Category UC-702

Radionuclides in Sediments and Seawater at Rongelap Atoll

V.E. Noshkin
W.L. Robison
R.J. Eagle
J.L. Brunk

Manuscript date: March 1998

LAWRENCE LIVERMORE NATIONAL LABORATORY
University of California • Livermore, California • 94551

Contents

List of Figures

List of Tables

Radionuclides in Sediments and Seawater at Rongelap Atoll

Victor E. Noshkin, William L. Robison, Rodney J. Eagle, James L. Brunk

Abstract. *The present concentrations and distributions of long-lived, man-made radionuclides in Rongelap Atoll lagoon surface sediments, based on samples collected and analyzed in 1981, are summarized in this report.*

The radionuclides were associated with debris generated with the 1954 Bravo thermonuclear test at Bikini Atoll. Presently, only ^{90}Sr and the transuranic radionuclides are found associated with the surface sediments in any quantity. Other radionuclides, including ^{60}Co and ^{137}Cs, are virtually absent and have either decayed or migrated from the deposits to the overlying seawater. Present inventories of ^{241}Am and $^{239+240}Pu$ in the surface layer at Rongelap are estimated to be 3% of the respective inventories in surface sediments from Bikini Atoll. There is a continuous slow release of the transuranics from the sediments back to the water column. The inventories will only slowly change with time unless the chemical-physical processes that now regulate this release to the water column are changed or altered.

Introduction

The current concentrations and distributions of long-lived, man-made radionuclides in Rongelap Atoll lagoon surface sediments, based on samples collected and analyzed in 1981, are summarized in this report. Rongelap Atoll was contaminated with close-in fallout from the 15-MT "Bravo" thermonuclear event that was detonated at Bikini Atoll on March 1, 1954. This test was the principal source for radioactive contamination now present in the environment. All other nuclear detonations during the 1956 Redwing and 1958 Hardtack series at the Pacific Proving Grounds contributed close-in fallout to Rongelap that amounted to a fraction of 1% of the 1954 amount, as determined from changes measured in gamma dose rates (Held, 1965). The historical events, and subsequent medical and some radiological surveillance studies associated with the fallout, the people, and the environment at Rongelap have been discussed in a number of reports and, most recently, in a series of articles which appeared in the July 1997 issue of *Health Physics*. These latter articles make reference to most, if not all, previous work at the atoll.

During the past 20 years, radiological surveys have focused on documenting levels of specific radionuclides in local foods, drinking water, and soils from islands of the atoll (Noshkin et al., 1981a; Robison et al., 1994; Robison and Conrado, 1996a,b; Robison et al., 1997), and in fish and invertebrates from the lagoon (Robison et al., 1981; Noshkin et al., 1981b). These surveys were undertaken to generate data to assess any potential radiological dose to present day and future inhabitants from external exposure, inhalation, and ingestion pathways.

Little information, however, is available on the man-made radionuclides in the lagoon sediments (and overlying seawater) that occupy the largest area of the atoll. Held (1963) provided a qualitative summary of early radiological data in some sediment, seawater, and other samples collected during the late 1950s. For the most part, only gross activity measurements were made on these early samples. However, in some samples collected up to 5 years after Bravo, concentrations of the fission products that included Zirconium-95

(^{95}Zr), Ruthenium-106 (^{106}Ru), Strontium-90 (^{90}Sr), Cesium-137 (^{137}Cs), Cerium-144 (^{144}Ce), Europium-155 (^{155}Eu), and Antimony-125 (^{125}Sb), and activation products that included Zinc-65 (^{65}Zn), Manganese-54 (^{54}Mn), Cobalt-57,60 (^{57,60}Co) , and Iron-55 (^{55}Fe), were separated and identified. The radionuclides contributing to the total activity in the sediments were associated mainly with the fine fraction (less than 0.14 mm) and were concentrated in the surface 5 to 10 cm. No man-made radionuclides were detected in seawater, but Held (1963) indicates that more sensitive analytical techniques undoubtedly would have revealed their presence.

The radionuclides in sediment and seawater do not contribute directly to human exposure. Because criteria related to human health is of primary concern, information on the fallout radioactivity in the lagoon sediments would be of secondary and lesser priority. However, Cohn et al. (1960) indicates the radionuclides are accumulated from seawater and sediments at Rongelap by local marine organisms that may eventually serve as food. Knowledge of the concentrations and distribution are required if the sediments are ever considered a resource to replace contaminated soil on islands of the atoll where they could then contribute to external exposure, and be available for uptake by indigenous plants and animals used as food. Finally, information on concentrations and distributions in sediments is of considerable scientific interest to better understand the behavior of the radionuclides within the aquatic environment of this atoll. These concerns justify an effort to learn about the radionuclides associated with the lagoon sediments and seawater.

During the 1978 Northern Marshall Island Survey (Robison et al., 1997), a few surface sediment and seawater samples were obtained for analysis from shallow depths on the lagoon reef to provide preliminary information on types and concentrations of radionuclides associated with the seawater and sediments (Noshkin et al., 1987). A more detailed sampling program was undertaken at Rongelap in 1981 to better define the distribution of persistent gamma-emitting radionuclides in the surface sediments. Lagoon seawater was again collected from selected stations in the lagoon. The results for the transuranic and other gamma-emitting radionuclides detected in the bottom sediments and in seawater are discussed in this report.

Experimental and Analytic Methods

Collection and Processing Methods

Nearshore sediment, lagoon seawater, fish, and invertebrates were collected from Rongelap Atoll in September 1978 during the Northern Marshall Islands Radiological Survey (NMRIS). Sediment was sampled to a depth of 4 cm from regions of the reef near fishing sites. Overlying water depths at the sampling locations ranged from 1 to 2 meters. Information describing the samples and sampling locations is provided in Table 1. Radionuclide concentrations in parts of fish and invertebrates collected at Rongelap during the survey are discussed in Noshkin et al. (1981a), and Robison et al. (1981).

During February 1981, sediment samples were collected at stations throughout the lagoon utilizing the Marshall Island research vessel, RV Liktanur. A portable winch, mounted on the deck of the ship, was used to lower and raise a Shipek grab sampler. Sampling locations were preselected using a systematic sampling plan and treating the atoll as a square grid to ensure all regions of the lagoon were sampled. Stations were separated approximately by two nautical miles so that each sampling site was the node of a 2 × 2 nautical mile grid. We deviated from this grid only when the overlying water depth at a station was too shallow for the research vessel, or when the bottom material was too difficult to sample. Depths of overlying water at the stations ranged from 12 to 56 meters. Three, 5.56-cm-diameter, circular core tubes were used to subsample the contents of each grab sample to a depth of 4.0 cm. The three samples were extruded from the cores into a single plastic bag. A total of 66 lagoon locations were sampled in this manner to generate a series of dimensionally comparative surface sediment samples. Our equipment limited us to sampling only the sediment surface. Sediments from depths deeper than 4 cm were not obtained. All sediment from the 1978 and 1981 programs was frozen and returned in a frozen state to LLNL for processing and analysis. At the laboratory, the

Table 1. Rongelap Atoll lagoon bottom surface sediments (0–4 cm) collected during September 1978 and February 1981.

Sample ID	Collection date		Latitude minutes N of 11° N	Longitude minutes E of 166° E	Water depth (m)	Area-72.75 cm^2 $g\ cm^{-2}$ dry wt
8123	9/23/78	1F	27	41.5	1	4.42
8122	9/23/78	7F	26	55	1	3.99
8119	9/22/78	9F	27	63.3	1	3.21
8120	9/22/78	13F	26	63.5	1	5.09
8121	9/22/78	23F	21.4	59.5	1	4.81
8117	9/20/78	33F	17	53.5	1	4.52
8116	9/20/78	42F	9	53	1	4.29
8118	9/20/78	42F	9	53	1	3.24
8115	9/19/78	46F	12	43	1	6.11
8114	9/18/78	47F	12.3	41.3	1	5.04
MSA554	2/16/81	–1	9.2	53.3	12	3.81
MSA555	–2		11	52	47	3.05
MSA556	–3		13	50	51	5.33
MSA557	–4		13	52	49	3.90
MSA558	–5		15	52	51	3.60
MSA559	2/17/81	–1	17	52	51	2.25
MSA560	–2		15	50	49	2.53
MSA561	–3		15	48	51	1.87
MSA562	–4		17	48	54	2.17
MSA563	–5		17	50	47	1.59
MSA564	–6		19	50	51	1.62
MSA565	–7		19	52	54	3.57
MSA566	–8		19	54	34	4.52
MSA567	–9		21	54	57	2.05
MSA568	–10		23	54	40	4.84
MSA569	–11		23	56	42	4.11
MSA586	–12		21	56	29	5.25
MSA587	–13		21	52	49	2.52
MSA588	–14		21	50	54	2.47
MSA589	–15		21	48	56	2.65
MSA590	–16		19	48	54	1.91
MSA591	2/18/81	–1	22	65	18	4.11
MSA592	–2		23	58	34	5.40
MSA593	–3		25	58	38	3.56
MSA594	–4		25	60	36	4.26
MSA595	–5		23	52	43	3.05
MSA596	–6		23	50	49	6.20
MSA597	–7		23	48	47	3.56
MSA598	–8		23	46	48	2.25
MSA599	–9		23	44	49	3.95
MSA600	–10		23	42	47	5.13
MSA611	2/19/81	–1	25	56	34	3.18

Table 1. (Continued).

Sample ID	Collection date	Latitude minutes N of 11° N	Longitude minutes E of 166° E	Water depth (m)	Area-72.75 cm^2 g cm^{-2} dry wt
MSA612	–2	25	54	44	3.66
MSA613	–3	25	52	47	3.55
MSA614	–4	27	52	34	1.00
MSA615	–5	27	50	42	4.08
MSA616	–6	25	50	48	3.45
MSA617	–7	25	48	46	3.31
MSA618	–8	25	46	51	2.60
MSA619	–9	25	44	47	3.75
MSA620	–10	25	42	46	4.71
MSA621	–11	21	42	46	3.22
MSA622	–12	21	44	54	5.61
MSA623	–13	21	46	55	4.82
MSA633	2/20/81 –1	19	46	51	1.55
MSA634	–2	19	44	49	1.53
MSA635	–3	19	42	46	0.71
MSA636	–4	19	40	42	5.40
MSA637	–5	17	40	42	5.88
MSA638	–6	15	40	40	4.00
MSA639	–7	13	40	29	5.66
MSA640	–8	13	42	35	5.69
MSA641	–9	13	44	40	5.75
MSA642	–10	15	46	48	2.21
MSA643	–11	15	44	49	5.25
MSA644	–12	15	42	46	5.14
MSA645	–13	17	42	47	3.95
MSA646	–14	17	44	44	5.61
MSA647	–15	17	46	55	1.65
MSA662	2/21/81 –1	13	46	44	5.29
MSA663	–2	13	48	48	5.66
MSA664	–3	11	48	37	5.11
MSA665	–4	10	48	35	5.33
MSA666	–5	11	46	40	5.42
MSA667	–6	11	50	42	5.39
MSA668	–7	10	50	33	5.54

sediment samples were thawed, and the wet and dry weights were determined. The sampling locations and other relevant information for the 1981 samples are listed in Table 1.

Radionuclide Analysis

Each sample was ball-milled and transferred to containers for analysis by gamma spectrometry using Ge(Li) detection systems. Counting times were usually 1000 minutes or longer for each sample. A general-purpose computer program was used for the data reduction of all gamma-ray spectra. A brief description of the gamma-ray program and an account of our quality assurance effort are given in Noshkin et al. (1988).

Representative detection limits (Bq kg^{-1}) for several gamma-emitting radionuclides in the 1981 samples were 1.0 for ^{60}Co, 0.9 for ^{137}Cs, 0.7 for Bismuth-207 (^{207}Bi), 3 for ^{125}Sb and ^{155}Eu, and 4 for Americium-241 (^{241}Am). All concentration data are reported on the date of collection except where noted. The radiological results for the 1978 and 1981 surface sediment samples are given in Tables 2 and 3.

Table 2. Concentrations of radionuclides in Rongelap surface sediments collected in September 1978.

Sample ID	^{241}Am Bq kg^{-1} dry wt	^{241}Am kBq m^{-2}	Error (%)	^{241}Am/$^{239+240}Pu$[a]	S Value[a,b]	^{155}Eu[a] Bq kg^{-1} dry wt	^{137}Cs[a] Bq kg^{-1} dry wt	^{90}Sr[a] Bq kg^{-1} dry wt
8123	11.0	0.49	4	0.63 (6)	0.21 (30)	7.4 (8)	1.6 (18)	16.0 (4)
8122	10.8	0.43	4	0.68 (6)	0.15 (40)	5.6 (14)	<0.2	29.3 (4)
8119	5.3	0.17	18			3.2 (14)	<0.2	
8120	13.0	0.66	5	0.67 (8)	0.11 (40)	5.7 (7)	0.5 (34)	40.7 (3)
8121	7.5	0.36	5			5.4(7)	0.6 (36)	21.0 (3)
8117	3.9	0.17	7	0.90 (8)	<0.2	2.6 (16)	<0.2	2.3 (14)
8116	3.6	0.15	5	0.60 (8)	<0.4	2.4 (10)	<0.2	4.7 (8)
8118	3.4	0.11	6	0.56 (8)		2.8 (9)	0.3 (37)	4.1 (7)
8115	5.6	0.34	4	0.61 (7)	<0.3	3.4 (9)	<0.1	5.1 (7)
8114	4.4	0.22	4	0.74 (6)	<0.4	3.7 (32)	<0.2	5.6 (6)
			mean	0.67 ± 0.11				
				0.72 ± 0.12[c]				

[a] Value in parenthesis is the one sigma counting error expressed as the percentage of the value listed.
[b] S is a dimensionless value and is expressed as a percentage to describe the activity of ^{238}Pu relative to the total alpha-emitting isotopes of plutonium.
S = (^{238}Pu alpha activity)/(^{238}Pu + ^{239}Pu + ^{240}Pu alpha activity)) × 100.
[c] Estimated ratio in 1997 based on estimation of ^{241}Pu in Bikini sediment (Noshkin et al., 1997a).

Table 3. Concentrations of gamma-emitting radionuclides in Rongelap surface sediments collected in February 1981.

MSA No.	^{241}Am Bq kg^{-1} dry wt	^{241}Am kBq m^{-2}	Error[a] (%)	^{155}Eu Bq kg^{-1} dry wt	^{155}Eu kBq m^{-2}	Error[a] (%)	Others detected[b] Bq kg^{-1} (% error)
554	3.2	0.12	100	3.6	0.14	100	
555	10.1	0.31	29	3.8	0.12	32	
556	3.3	0.18	100	2.4	0.13	100	
557	5.6	0.22	19	3.0	0.12	34	
558	4.1	0.15	100	2.6	0.09	100	
559	32.7	0.74	12	11.5	0.26	16	
560	21.5	0.54	16	6.1	0.16	22	
561	18.6	0.35	29	3.0	0.06	100	
562	13.6	0.30	27	4.2	0.09	53	
563	13.9	0.22	15	4.9	0.08	32	
564	25.9	0.42	16	1.5	0.02	100	
565	12.5	0.45	11	4.1	0.15	24	
566	3.7	0.17	100	2.6	0.12	100	
567	20.0	0.41	16	6.5	0.13	28	
568	16.6	0.80	23	4.6	0.22	38	^{60}Co–1.7(57)
569	9.7	0.40	32	7.7	0.32	30	
586	4.4	0.23	100	3.0	0.16	100	
587	24.8	0.62	14	6.7	0.17	31	
588	29.8	0.74	12	8.4	0.21	18	
589	21.1	0.56	23	10.7	0.28	19	
590	23.3	0.45	19	6.8	0.13	38	^{40}K–42(39)
591	8.9	0.37	22	4.2	0.17	38	
592	3.7	0.20	100	2.6	0.14	100	
593	26.3	0.94	10	5.9	0.21	22	
594	15.0	0.64	24	3.0	0.13	100	
595	41.5	1.27	9	15.9	0.49	13	
596	23.3	1.45	18	5.9	0.37	29	
597	30.4	1.08	16	12.6	0.45	16	^{137}Cs–1.4(38); ^{40}K–21(31)
598	41.1	0.93	14	12.6	0.28	17	^{137}Cs–2.4(38); ^{40}K–37(36)
599	25.6	1.01	12	8.7	0.34	14	
600	21.5	1.10	18	8.4	0.43	18	
611	17.1	0.54	21	3.3	0.11	100	
612	23.6	0.86	14	9.0	0.33	18	
613	15.2	0.54	25	3.0	0.11	100	
614	32.6	0.33	20	8.7	0.09	47	
615	24.3	0.99	10	9.7	0.40	15	
616	38.1	1.32	13	14.1	0.49	12	^{137}Cs–2.3(25)
617	32.8	1.09	6	12.0	0.40	11	^{137}Cs–1.7(41)
618	47.5	1.23	17	17.5	0.45	15	^{137}Cs–2.6(28)

Table 3. (Continued).

MSA No.	^{241}Am Bq kg^{-1} dry wt	^{241}Am kBq m^{-2}	Error[a] (%)	^{155}Eu Bq kg^{-1} dry wt	^{155}Eu kBq m^{-2}	Error[a] (%)	Others detected[b] Bq kg^{-1} (% error)
619	33.1	1.24	10	9.4	0.35	16	
620	41.5	1.96	15	14.0	0.66	18	^{137}Cs–2.5(37)
621	15.8	0.51	17	8.8	0.28	23	
622	25.5	1.43	17	8.3	0.47	17	^{40}K–17(31)
623	28.3	1.37	7	9.3	0.45	13	
633	27.7	0.43	15	9.6	0.15	26	
634	24.2	0.37	22	8.2	0.13	25	
635	28.4	0.20	8	10.7	0.08	17	
636	10.7	0.58	35	2.8	0.15	100	
637	13.2	0.78	26	4.1	0.24	36	
638	7.1	0.29	40	4.6	0.18	28	
639	3.1	0.17	100	3.5	0.20	100	
640	2.9	0.16	100	2.1	0.12	100	
641	7.3	0.42	22	3.4	0.20	32	
642	16.8	0.37	21	3.9	0.09	51	
643	13.0	0.68	25	4.6	0.24	32	
644	4.9	0.25	100	3.4	0.18	100	
645	15.6	0.62	20	3.5	0.14	100	^{137}Cs-2.8(23); ^{40}K-15(40)
646	7.2	0.40	36	2.6	0.14	100	
647	17.6	0.29	12	4.7	0.08	22	
662	4.1	0.22	100	3.1	0.16	100	
663	5.3	0.30	100	3.6	0.20	100	
664	3.5	0.18	100	3.9	0.20	100	
665	2.5	0.13	100	2.1	0.11	100	
666	3.6	0.20	100	4.4	0.24	38	^{137}Cs-2.0(33)
667	4.7	0.25	26	3.1	0.17	52	
668	3.8	0.21	100	2.7	0.15	100	

[a] Error(%)—one sigma counting error expressed as the percentage of the value listed.
[b] Except where indicated, ^{60}Co was below detection limits of 1.0 ± 0.5 Bq kg^{-1} in remaining samples.
[b] Except where indicated, ^{137}Cs was below detection limits of 0.9 ± 0.3 Bq kg^{-1} in remaining samples.
[b] ^{207}Bi was below detection limits of 0. 7 ± 0.3 Bq kg^{-1} in all samples.

The concentrations of Plutonium-239+240 ($^{239+240}Pu$) and ^{90}Sr were determined in the 1978 samples following chemical separation procedures described in Wong et al. (1994). Plutonium-239+240 and ^{90}Sr were not separated from the 1981 samples. Several radionuclides were separated and measured in the lagoon seawater samples collected during 1978 and 1981. Water samples were first filtered through an in-line filter. The filter used to remove particulates from the samples is a 1–µm Micro-Wyndell®, DDCCPY filter cartridge (AMF, Cuno Division, Meriden, Connecticut). We have demonstrated that this filter is as efficient in removing suspended particulates from the lagoon water as a 0.2 to 0.3–µm filter (Wong et al., 1980). Therefore, solution concentrations shown in Table 4 refer to

Table 4. Concentration of radionuclides in surface seawater from Rongelap Atoll during September 1978 and February 1981.

Island ID	Latitude minutes N of 11° N	Longitude minutes E of 166° E	$^{239+240}Pu$ solution $mBq\,l^{-1}$	Error[a] (%)	$^{239+240}Pu$ prefilter $mBq\,l^{-1}$	Error[a] (%)	^{241}Am solution $mBq\,l^{-1}$	Error[a] (%)	^{241}Am prefilter $mBq\,l^{-1}$	Error[a] (%)	^{137}Cs $mBq\,l^{-1}$	Error[a] (%)	^{90}Sr $mBq\,l^{-1}$	Error[a] (%)
Rongelap Atoll (September 1978)														
F-1	27	41.5	57	12			12	28	10	26	4.96	3	4.11	6
F-7	26	55	42	22			29	32	7	39	5.00	3	4.11	10
F-9	27	63.3	39	14	106	5					4.96	3		
F-23	21.4	59.5	46	22	26	17	11	50	18	13	6.96	3	4.37	4
F-33	17	53.5	64	11	4	36	28	34	4	57	5.70	3	3.67	7
F-47	12.3	41.3	42	31	62	24	7	100	11	50	4.78	7	4.15	11
		Mean	48 ± 10								5.4 ± 0.8		4.1 ± 0.3	
Rongelap (February 1981)														
D758	13	50	52	8										
D759	9	52	44	9										
D760	19	50	41	13										
D761	17	53.3	81	7										
D762	26	32	30	12										
D763	26	55	25	16										
D764	25	56	20	14										
D765	19	40	33	12										
D766	13	48	41	12										
D767	26.7	41	44	12										
D768	10	50	41	10										
		mean	41 ± 16											
Mean concentration in north equatorial Pacific surface water 1972–1984			15								5.2 ± 0.4			

[a] Error (%)—one sigma counting error expressed as the percentage of value listed.

the quantity passing through a 1–μm filter and prefilter samples refer to the particulate phase associated with the 1–μm filter.

Discussion

Radionuclides Associated with Sediments 24–27 Years Post Bravo

The data in Tables 2 and 3 show that the man-made gamma-emitting radionuclides found above detection limits in most 1978 and 1981 samples included only ^{241}Am and ^{155}Eu. By 1981, ^{137}Cs was identified in only 8 of the 66 samples at a level no greater than 3 Bq kg^{-1} and ^{60}Co was detected in only one sample. In a few samples, naturally occurring Potassium-40 (^{40}K), and daughter products in the uranium decay series (concentrations not provided in the tables) were the only other gamma-emitting radionuclides detected with the Ge(Li) systems. Other man-made gamma-emitting radionuclides, such as ^{207}Bi, $^{152,154}Eu$, ^{125}Sb, and Rhodium-101,102m ($^{101,102m}Rh$), measured in some sediment samples during the same period from Bikini lagoon (Noshkin et al., 1997b), were below detection limits in all Rongelap samples. Other man-made, gamma-emitting radionuclides, that were shown by Held (1963) to be associated with the sediments in the late 1950s have been reduced to levels that are below detection limits in 100–300-g samples.

In 1981, the mean ^{155}Eu to ^{241}Am activity ratio in surface sediments, computed from the data in Table 3, was 0.37 ± 0.12. By 1997, the ratio is reduced to at least 0.040 ± 0.012 due to loss of ^{155}Eu by radioactive decay. It will essentially be below detection limits in any surface sediment sample collected after 1997. Of the many gamma-emitting radionuclides originally deposited in the lagoon with fallout debris, only ^{241}Am will be measurable at concentrations not exceeding 50 Bq kg^{-1} dry weight in lagoon surface sediments through the year 2000. In island soils, however, mean levels of ^{137}Cs presently exceed those of ^{241}Am (Robison and Conrado, 1996a,b). Therefore, ^{137}Cs will be measurable through the year 2000, but only in association with materials originating from the terrestrial environment.

In 1978, the mean ^{241}Am to $^{239+240}$Pu activity ratio in the surface sediments from eight different locations was 0.67 ± 0.11. This value is assumed to be representative of the ratio in sediments from anywhere within the atoll. It is in excellent agreement with the mean value of 0.69 ± 0.17 determined in 19 surface sediment samples collected at Bikini Atoll in 1979 (Noshkin et al., 1997a). The ratio will increase slightly to a value of 0.72 ± 0.12 by 1997 from growth of ^{241}Am based on estimated residual quantities of the parent radionuclide, ^{241}Pu, in the environment (Hisamatsu and Sakanoue, 1978; Noshkin et al., 1997a). Therefore, $^{239+240}$Pu will be measurable in the sediments through the year 2000 at levels somewhat higher than ^{241}Am.

The S value, which describes the activity of ^{238}Pu relative to the total alpha-emitting isotopes (^{238}Pu + ^{239}Pu + ^{240}Pu) as a percent, was computed in only three samples where ^{238}Pu was above detection limits. These three samples were obtained from the northern half of the lagoon. The mean S value associated with the samples is 0.16 ± 0.05. This is comparable to an S value of 0.10 ± 0.01, determined at this laboratory, in "Bravo" test debris deposited on the fishing boat, Fukuryu-Maru. The boat was located east of Bikini and to the north of Rongelap atoll at 166 degrees 58 minutes N, 11 degrees 53 minutes E on March 1, 1954, and was contaminated with fallout debris from the Bravo test. The similar mean S value associated with the fallout debris from the ship and in the sediment from the atoll indicates, as expected, a common source for this radioactivity. In 1997, the S value will be reduced slightly to 0.09 due to decay of ^{238}Pu. Concentrations of ^{238}Pu in this environment are of little radiological significance.

Strontium-90 was also detected in the 1978 lagoon sediment samples following radiochemical separation and analysis of its daughter radionuclide, Yttrium-90 (^{90}Y), by beta counting. This radionuclide is strongly associated with carbonate sediments and will slowly decrease in the surface sediments primarily by radioactive decay. Unlike ^{155}Eu and $^{239+240}$Pu, ^{90}Sr and ^{241}Am are not strongly correlated in the sediments. Concentration ratios of ^{241}Am to ^{90}Sr are found to range from 0.3 to 1.7, respectively.

Neither ^{241}Am, $^{239+240}$Pu, nor ^{90}Sr are significantly concentrated in edible parts of marine organisms. These radionuclides in any marine food chain will continue to contribute only a small fraction of the total effective dose to individuals at this atoll (Robison, et al., 1981; Robison et al., 1994).

Distribution of ^{241}Am in the Surface Sediments

The surface sediment inventory values for ^{241}Am were plotted on a lagoon chart. Isoconcentration lines were constructed to distinguish regions of the bottom surface sediment with comparable values. The data provided in Table 3 were used to construct the spatial distribution shown in Figure 1. Highest inventories are associated with the sediment from the northern half of the atoll. Gradients decrease in a southerly direction to regions near the South Pass where ^{241}Am is below detection in the surface sediments. The different isopleths roughly coincide with the estimated fallout path of differently contaminated debris from the 1954 Bravo event (Robison et al., 1997). There is approximately a factor of 10 difference in concentration associated with surface sediments from the north compared to samples from the southern part of the atoll. This was also the difference estimated for the gamma dose rates on northern and southern islands one day after the Bravo test (Held, 1965). Following deposition to the lagoon surface water, some fraction of the particles contaminated with ^{241}Am (or its parent, ^{241}Pu)

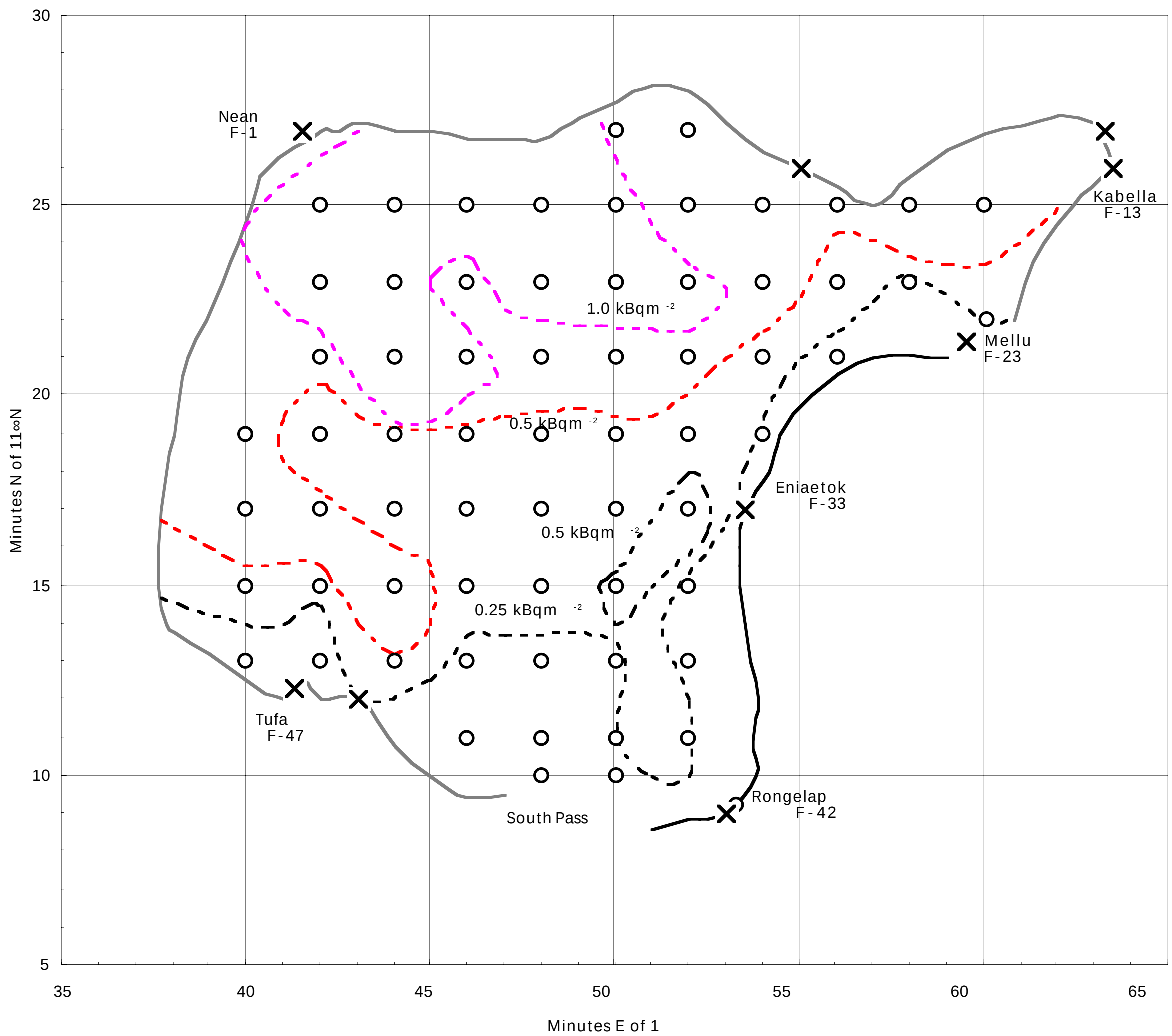

Figure 1. Isoconcentration plot of the inventory for ^{241}Am (kBq m^{-2}) in the surface (0–4 cm) sediment from Rongelap lagoon.

settled rapidly to the bottom sediment, which accounts for the current distribution pattern that resembles the original 1954 fallout pattern over the atoll.

In Figure 1, the area between the contour intervals was determined along with the lognormal mean inventory (kBq m^{-2}) within the respective regions. Multiplying the area by the mean inventory provides an estimate of the total amount of ^{241}Am associated with the sediment within the region. Summing these regional amounts generates a value for the total amount associated with the surface
4-cm layer of sediment over the entire lagoon. The mean activity ratio for ^{241}Am to $^{239+240}$Pu of 0.67 ± 0.11 is used to estimate the amount of $^{239+240}$Pu associated with the surface sediments. Respective areas and inventory values are shown in Table 5. The mean concentrations (Bq kg^{-1}) within the inventory intervals are also calculated and shown in Table 5 for information. The total quantities of ^{241}Am and $^{239+240}$Pu in the lagoon-sediment surface (4–cm deep) of are 0.63 ± 0.09 and 0.94 ± 0.16 TBq,

Table 5. Inventory and concentration of ^{241}Am and $^{239+240}$Pu in surface sediments from regions of Rongelap lagoon as of February 1981. Area of Rongelap lagoon—1025 km^2.

^{241}Am Inventory interval (kBq m^{-2})	Mean inventory (kBq m^{-2})	^{241}Am Mean concentration (Bq kg^{-1})	Area of interval (km^2)	TBq in interval to depth of 4 cm
>1	1.3 ± 0.3	31 ± 10	204	0.27 ± 0.06
0.5-1	0.7 ± 0.2	20 ± 9	289	0.20 ±0.06
0.25–0.5	0.36 ± 0.10	16 ± 8	333	0.12 ± 0.03
0-0.25	0.19 ± 0.03	5 ±2	199	0.04 ± 0.01
			Lagoon total	0.63 ± 0.09

Estimated inventory of $^{239+240}$Pu in surface sediments from regions of Rongelap lagoon

^{241}Am inventory interval (kBq m^{-2})		TBq $^{239+240}$Pu in interval to depth of 4.0 cm[a]
>1		0.40 ± 0.11
0.5-1		0.30 ± 0.10
0.25-0.5		0.18 ± 0.05
0-0.25		0.06 ± 0.02
	Lagoon total	0.94 ± 0.16

[a] ^{241}Am-inventory value divided by 0.67 ± 0.11.

respectively. These quantities represent approximately 3% of the respective inventories of ^{241}Am and $^{239+240}$Pu associated with the surface sediment (4 cm deep) at Bikini Atoll (Noshkin et al., 1997a). The highest levels of ^{241}Am in surface sediments found in the NW quadrant of Rongelap are equivalent in value to the lowest levels in surface material found near the East Channel of Bikini lagoon. Unlike Bikini, the surface sediments from anywhere within Rongelap lagoon contain essentially no ^{137}Cs and low levels of the transuranic radionuclides. The sediments are less contaminated than the soil from the islands, and consequently, provide a better carbonate material if replacement of contaminated island soil is required at either Bikini or Rongelap Atolls. These estimated inventories must be viewed as lower limits since the radionuclides are no doubt present in material from greater depths in the sediment column. At Bikini, for example, only 28 ± 10% of the ^{241}Am is associated with the surface 4 cm of sediment, and recall that Held (1965) indicates that early sediment samples contained radioactivity to at least a depth of 10 cm. At this time, it is not possible to speculate on the fraction of the total inventory represented by the amount in the surface 4 cm layer of sediment.

Concentrations in Seawater

The data in Table 4 shows that the concentrations of $^{239+240}$Pu in all lagoon water collected in 1978 and 1981 exceed the background levels of 15 ± 7 µBq l^{-1} measured in the equatorial Pacific surface waters between 1972 and 1982 (Noshkin et al, 1987). The comparable mean amounts of $^{239+240}$Pu found in solution during both years shows that these higher than oceanic background levels are real and persistent. Mobilization of small amounts of $^{239+240}$Pu (and ^{241}Am) from sediments to the overlying bottom water is responsible for the elevated levels in the seawater within Rongelap lagoon. A comparable process adds excess transuranic radionuclides to the water column at Bikini and Enewetak (Noshkin, 1980; Noshkin and Wong, 1980; Noshkin et al., 1987).

Data in Table 4 show that the average concentration of ^{137}Cs in surface equatorial seawater between 1967 and 1982 was 5.2 ± 0.4 mBq l^{-1} (Noshkin et al., 1987). Unlike the $^{239+240}$Pu and ^{241}Am concentrations, the ^{137}Cs levels in the lagoon seawater are indistinguishable from fallout levels in the equatorial Pacific Ocean. Essentially the entire inventory of ^{137}Cs associated with the local fallout that reached the lagoon sediments in 1954 has mobilized to the overlying seawater. This labeled water is eventually mixed with surface water that is then transported by prevailing surface currents out of the lagoon to the surface water of the north equatorial Pacific ocean.

Concentrations of ^{90}Sr in the lagoon water are not significantly different from the global fallout concentrations in lagoon seawater at less contaminated atolls such as Ujelang, Likiep, and Wotho (Noshkin et al., 1987). Therefore, unlike $^{239+240}$Pu and ^{241}Am, any ^{90}Sr mobilized from sediments to seawater is masked by the throughput of global fallout concentrations associated with the north equatorial surface water, which continuously exchanges with the lagoon water mass.

Conclusions

The lagoon sediments at Rongelap Atoll were contaminated with a variety of fission and activation products from settling fallout particles generated by the Bravo thermonuclear test held at Bikini Atoll in 1954. Other tests conducted during 1956 and 1958 at Bikini and Enewetak added less than a fraction of 1% of that from the Bravo event to the close-in fallout at Rongelap. By 1997, the only long-lived radionuclides found in any quantity associated with lagoon surface sediments at Rongelap Atoll would be ^{90}Sr and the transuranic radionuclides, ^{241}Am and $^{239+240}$Pu. These radionuclides are not significantly concentrated in edible parts of marine organisms, and will continue to contribute only a small fraction of the total effective dose to individuals at this atoll from the marine ingestion pathway. Highest amounts of ^{241}Am are associated with bottom sediments from the northern half of the atoll. Concentration gradients decrease in a southerly direction to regions near the South Pass where ^{241}Am is below limits of detection. Little post-depositional migration of particles with ^{241}Am contamination appears to have occurred since the fallout material fell to the bottom of the lagoon in 1954. The inventories of ^{241}Am and $^{239+240}$Pu in the surface 4 cm of sediment are approximately 3% of the quantity in surface sediments from Bikini Atoll. Since the sediments contain no measurably high levels of other gamma emitters and low levels of the transuranic radionuclides, they are radiologically better materials if sediments are ever required to replace contaminated soil on either Bikini or Rongelap Atolls. There is a continuous low release of $^{239+240}$Pu and ^{241}Am from the sediments back to the water column. The inventories will change only slowly with time unless the chemical/physical processes that now regulate this release to the water column are changed or altered.

References

Cohn, S. H., J. S. Robertson, and R.A. Conard (1960), "Radioisotopes and Environmental Circumstances: The Internal Radioactive Contamination of a Pacific Island Community Exposed to Local Fallout," in Radioisotopes in the Biosphere, Caldecott, R. S., and Snyder, L. A., Eds., University of Minnesota Printing Department, Minneapolis, MN.

Held, E. E. (1963), "Qualitative Distribution of Radionuclides at Rongelap Atoll," in Radioecology, Schultz, V., and Klement, A.W. Jr., Eds., Reinhold, New York, NY.

Held, E. E. (1965), Gamma Dose Rates at Rongelap Atoll, 1954-1963, University of Washington, Laboratory of Radiation Biology, Seattle, WA, UWFL-91.

Hisamatsu, S., and M. Sakanoue (1978), "Determination of Transuranium Elements in a So-Called "Bikini Ash" Sample and in Marine Sediment Samples Collected Near Bikini Atoll," Health Phys. 35, 301–307.

Noshkin, V. E. (1980), "Transuranium Radionuclides in Components of the Benthic Environment of Enewetak Atoll," in Transuranic Elements in the Environment, Hanso, W. C., Ed., U.S. Department of Energy, Washington, DC, DOE/TIC-22800.

Noshkin, V. E., and K. M. Wong (1980), "Plutonium Mobilization from Sedimentary Sources to Solution in the Marine Environment," Marine Radioecology (Proc. 3rd NEA Seminar, Tokyo, 1979), NEA, OECD, Paris, p.729.

Noshkin, V. E., K. M. Wong, R. J. Eagle, and W. L. Robison (1987), "Comparative Concentrations of 137Cs, 90Sr, 239,240Pu and 241Am in Tissues of Fish from the Marshall Islands and Calculated Dose Commitments from Their Consumption," in Environmental Research on Actinide Elements, Pinder, J .E. III, Alberts, J. J., McLeod, K. W., and R. G. Schreckhise, Eds., Proc. Symposium held at Hilton Head, SC, 1983, U.S. Department of Energy, Washington, D.C., CONF-841142.

Noshkin, V. E., K. M. Wong, R. J. Eagle, T. A. Jokela, and J. A. Brunk (1988), Radionuclide Concentrations in Fish and Invertebrates from Bikini Atoll, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-53846.

Noshkin, V. E., R. J. Eagle, K. M. Wong, and W. L. Robison (1997a), Sediment Studies at Bikini Atoll Part II. Transuranium Elements in Surface Sediments, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-LR-129379.

Noshkin, V. E., R. J. Eagle, J. L. Brunk, and W. L. Robison (1997b), Sediment Studies at Bikini Atoll Part II1. Inventories of Some Long Lived Gamma Emitting Radionuclides Associated with Lagoon Surface Sediment, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-LR-129834.

Noshkin, V. E., R. J. Eagle, K. M. Wong, T. A. Jokela, and W. L. Robison (1981a), Radionuclide Concentrations and Dose Assessment of Cistern Water and Groundwater at the Marshall Islands, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-52853 Pt. 2.

Noshkin, V. E., R. J. Eagle, K. M. Wong, T. A. Jokela, J. L. Brunk, and K. V. Marsh (1981b), Concentrations of Radionuclides in Reef and Lagoon Pelagic Fish from the Marshall Islands, Lawrence Livermore National Laboratory, Livermore, CA, UCID-19028.

Robison, W. L., and C. L. Conrado (1996a), Radiological Conditions at the Southern Islands of Rongelap Atoll, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-ID-123375.

Robison, W. L. and C. L. Conrado (1996b), Radiological Conditions at Naen, Yugui, Lomiulal, Kabelle, and Mellu Islands in the Northern Half of Rongelap Atoll, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-ID-123374.

Robison, W. L., C. L. Conrado, and K. T. Bogen (1994), An Updated Dose Assessment for Rongelap Island, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-LR-107036.

Robison, W. L., V. E. Noshkin, C. L. Conrado, R. J. Eagle, J. L. Brunk, T. A. Jokela, M. E. Mount, W. A. Phillips, A. C. Stoker, M. L. Stuart, and K. M. Wong (1997), "The Northern Marshall Islands Radiological Survey: Data and Dose Assessments," Health Phys. 73, 37–48.

Robison, W. L., V. E. Noshkin, W. A. Phillips and R. J. Eagle (1981), The Northern Marshall Islands Radiological Survey: Radionuclide Concentrations in Fish and Clams and Estimated Doses Via the Marine Pathway, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-52853 Pt. 3.

Wong, K. M., T. A. Jokela, and V .E. Noshkin (1994), Radiochemical Procedures for Analysis of Pu, Am, Cs, and Sr in water, Soil, Sediments and Biota Samples, Lawrence Livermore National Laboratory, Livermore, CA, UCRL-ID-116497.

Wong, K. M., T. A. Jokela, and V. E. Noshkin (1980), "Problems Associated with Transuranium Determination of Suspended Soilids in Sea Water Samples," in Radioelement Analysis Progress and Problems, Lyon, W.S., Ed., Ann Arbor Press, Ann Arbor, MI.